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Pseudo-ductile behaviour in fibre reinforced thermoplastic angle-ply composites

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A R T I C L E I N F O

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A B S T R A C T

Pseudo-ductile behaviour has previously been demonstrated in a $[\pm 45_n]_s$ carbon fibre composite *via* fibre rotation. To further develop the pseudo-ductile response, the high strain properties of the matrix should be considered to avoid strain localisation and potentially exploit strain-hardening in an amorphous thermoplastic matrix. For the first time, polycarbonate, a high strain-to-failure matrix is utilised in a pseudo-ductile composite design and compared to a high performance thermoset fibre composite containing an epoxy resin. The use of polycarbonate leads to enhanced pseudo-ductile strain (49%), longitudinal strength (24%) and apparent in-plane shear strength (26%), albeit with a reduction in elastic modulus (26%), shear chord modulus (22%) and yield strength (26%).

1. Introduction

True ductility can be defined by a material's ability to withstand load beyond the yield stress with minimal damage and to be re-loaded with no loss of modulus. This is commonly observed in metals, but carbon fibre reinforced composites typically fail catastrophically, often with little warning that the load limit has been reached [1]. Pseudo-ductile behaviour has been achieved in angle-ply laminates, and although that alleviates the brittleness of fibre-reinforced composites [2], it does not usually allow recovery when re-loaded. In effect, angle plies exploit additional length from the inextensible fibres by undergoing rotation under load.

In optimising the strength and modulus, the fibre angle with respect to the loading direction can be reduced at the cost of reducing strain-to-failure. In order to achieve a higher strain to failure, thin ply composites have successfully been employed [2], as premature damage, due to matrix cracking and delamination from high free-edge interlaminar stresses, can be suppressed with ply thicknesses of <0.06 mm for unidirectional composites. These thin plies are reported to lead to an increase in the delamination initiation stress [3]. At high strains, strain localisation could form, subsequently, the matrix warrants further investigation if the concept of pseudo-ductility through fibre rotation is to be exploited. Furthermore, given the capabilities of some amorphous and semi-crystalline polymers to undergo strain-hardening during large-strain plastic deformation, combining such a matrix with the angle-ply pseudo-ductile composite concept may offer attractive tensile properties.

Strain hardening is initiated in a polymer when it experiences such a level of stress, that the intermolecular attractions are unable to resist large-scale segmental motion; primary bonds inhibit further motion as a result of steric hindrance, leading to strain-hardening. This chain orientation is typically feasible in amorphous polymers, or in an amorphous region in a semi-crystalline polymer, and is significantly pronounced under uniaxial tensile loading as a result of large-scale polymer chain alignment parallel to the loading direction [4,5]. Additional strain hardening mechanisms are strain-induced re-crystallisation and crazing: the former occurs in semi-crystalline polymers and enables the formation of aligned structures adjacent to the aligned crystalline polymers; the latter occurs under tensile loading only and arises from the formation of voids that are connected by aligned polymer structures. Slipping amongst the aligned polymer structures can be an effective mechanism to increase toughness and improve the ductility of an amorphous

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polymer [6].

In this study, a thermoset and thermoplastic matrix were selected for consideration: the choice of an epoxy resin was straightforward as the difunctional monomer based on the diglycidyl ether of bisphenol A is commonly studied in the literature. When considering the choice of a suitable thermoplastic to employ, a less commonly used matrix, polycarbonate, was selected. Polypropylene and polyamide (6,6) are both used in various automotive composite applications and selected properties are compared in Table 1 (along with the epoxy and polycarbonate). Polypropylene is one of the more popular thermoplastics used in a fibre-reinforced form and it offers the advantages of short processing times, low density, corrosion resistance and quick repair, justifying its potential as a replacement for conventional automotive materials [7,8]. The engineering polymers of polycarbonate and polyamide (6,6), have superior mechanical properties to polypropylene and both are commercially available in reinforced format. Of these, polycarbonate displays the highest modulus with comparable tensile strength to polyamide (6,6), while displaying a significantly lower moisture absorption, which potentially limits the use of polyamides in conditions of high humidity or following frequent contact with water [9]. This combination of favourable properties makes the relatively limited uptake or study of carbon fibre reinforced polycarbonate surprising.

Polycarbonate is typically favoured as a polymer for its impact toughness and both thermal and dimensional stability, furthermore, its inherent ability of being recycled prompts investigation as a replacement for thermoset matrices. Polycarbonate has strain hardening potential and exhibits crazing [10], moreover, it has a significant strain-to-failure (90% [11]) compared to typical, aerospace grade thermoset epoxy resin (~3% [12]). At low strain rates ($<10\ s^{-1}$) polycarbonate demonstrates a reduction in yield stress and modulus, but strain hardening at low strain rates is more prominent with an increase in strain-to-failure [13]; this has been attributed to the activation of the β transitions at higher strain rates. Subsequently, this work builds on the progress made on pseudo-ductile angle ply thermoset composites [2], potentially applying the pseudo-ductile concept in areas where polycarbonate prevails as a high impact tough material.

Table 1 displays the physical and mechanical data for polycarbonate and an epoxy resin (Prime 20LV, Gurit), which has been used as a comparison in this work. Although both polymers display similar tensile strengths, polycarbonate offers inferior tensile modulus when compared with Prime 20LV epoxy resin; therefore, a reduction in mechanical performance, such as shear chord modulus is expected when the polymer is consolidated in an angle ply composite. The significantly contrasting strain-to-failure of the polymers tests the hypothesis that the high strain-to-failure of the polycarbonate assists in enhancing pseudo-ductile performance.

Table 1
Comparison of polycarbonate, (Prime 20LV) epoxy resin and polyamide (6,6) and polypropylene.

Property	Epoxy Resin (Prime 20LV -Slow hardener)	Polycarbonate	Polyamide (6,6)	Polypropylene
Density	1.10 [11]	1.20 [11,14]	1.13–1.15 [14]	0.9–0.91 [14]
T_g (°C)	83 [11]	145 [11]	48 [11]	−10 [11]
T_m (°C)	N/A	N/A	265 [11]	170 [11]
Water absorption, 24 h (%)	N/A	0.15 [11]	1.50 [11]	0.02 [15]
Tensile strength (MPa)	70.0 [11]	62.8–72.4 [11, 14]	75.9–94.5 [11,14]	31.0–41.4 [14]
Tensile modulus (GPa)	3.50 [11]	2.30–2.40 [14, 16]	1.58–1.38 [14]	1.14–1.55 [14]
Strain-to-fail (%)	3.5 [11]	90–150 [11, 14]	15–300 [14]	100–600 [14]

The aim of this study is to: (1) evaluate the pseudo-ductile response for a high strain-to-fail matrix, (2) exploit polycarbonate's potential strain-hardening behaviour in a pseudo-ductile angle ply composite design, and (3) determine the viability of utilising thermoplastics for pseudo-ductility in angle ply design as an alternative to thermoset matrices.

Subsequently, the in-plane shear response as a function of the matrix will be considered, by testing an amorphous polymer, polycarbonate, reinforced with woven carbon fibre plies [±45], and comparing to similarly fibre reinforced epoxy resin composites, for the first time.

2. Experimental methods

2.1. Sample fabrication and mechanical testing

Polycarbonate prepreg (ePreg range) was supplied by Engineered Cramer Composites (Heek, Germany). The prepreg contained sized-removed Torayca FT300B 3000-40B (3k) carbon fibres, which were used throughout this study, configured in a 5-harness satin (290 g/m^2 fibre areal weight) and laid up as $[\pm 45_5]_2$.

The same type of carbon fibres (sized-removed using a proprietary process) were also infused with a thermoset matrix (Prime20 LV with slow hardener) and cured following the manufacturer's recommended procedure of 7 h at 65 °C in an oven. For the PC composite, the pre-preg plies were laid up and consolidated in a heat press which had the capabilities to record load, pressure and temperature *via* thermocouples located in the press platens adjacent to the sample. The load, pressure and temperature profile recorded can be seen in the supplementary information, Fig. S1b and this can be compared to the manufacturer's recommended fabrication procedure in Fig. S1a. Hereinafter, the fibre reinforced epoxy resin and polycarbonate composites are referred to as ER and PC, respectively.

Three-point micrometer measurements of laminate thickness and width were performed prior to testing. Subsequently, the plies in this study had a final ply consolidation thickness of 0.318 mm for PC and 0.305 mm for ER, leading to an estimated fibre volume (V_f) of 51% for PC and 53% for ER.

An in-plane shear test was conducted by tension on the±45 laminates as per ASTM D3518 [17]. A schematic diagram displaying the location of the video gauge strain markers can be seen in Fig. 1. The tabs were E-glass pre-preg composite and bonded to the test coupons using 3M DP490 adhesive.

All mechanical tests were conducted using a hydraulic-actuated Instron test machine, under displacement control with a displacement rate of 2 mm/min, as per ASTM D3518 [17]. Longitudinal and transverse strains (see location of trackers in Fig. 1) were measured *via* an Imetrum Video gauge.

The in-plane shear stress was calculated using $\tau_{12}^i = \sigma_x^i/2$, where σ_x^i is the true longitudinal stress at *i*-th data point; superscript *m* refers to the maximum point or strength. The shear chord modulus was calculated

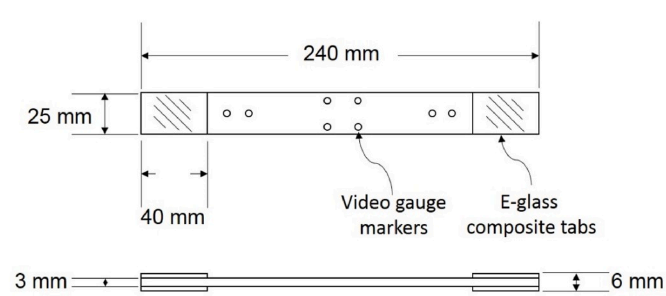

Fig. 1. Nominal sample dimensions with approximate locations of the video gauge sensor markers.

using $G_{12}^{chord} = \Delta\tau/\Delta\gamma$, where $\Delta\tau$ is the difference in applied true shear stress between two true shear strain points and $\Delta\gamma$ is the difference between two shear strain points. A shear strain range of 1500–5500 με was used, as recommended by the ASTM standard [18]. The shear strain (γ) was calculated using $\gamma = \varepsilon_{x,i} - \varepsilon_{y,i}$, where $\varepsilon_{x,i}$ is the true longitudinal strain and $\varepsilon_{y,i}$ is the true transverse strain at *i*-th data point.

2.2. Definition of yield and pseudo-ductility

Yield stress, $\sigma_{Yield,}$ and pseudo-ductile strain, ε_d, are shown graphically in Fig. 2. σ_{Yield} is defined as the point of intersection between the laminate stress-strain curve and a parallel straight line to the initial modulus with an offset of 0.1% strain. The pseudo-ductile strain, $_{d,}$ is the failure strain minus the strain for the concurrent stress parallel line to the initial modulus [2]. To differentiate terms, an asterisk has been used to denote 'to failure'.

3. Results

3.1. Mechanical testing

Five samples each for ER and PC were tested. The σ_x, plotted against $_x$ and $_y$, for ER (a) and PC (b) is shown in Fig. 3. As expected, there is a highly non-linear stress-strain behaviour for both matrices. Evidently, there is a period of linearity (to $_{x}$, $_{y}$ = ~0.2%), a yielding then followed by a large period of pseudo-ductility. Consequently, a high average strain-to-failure was observed for both ER and PC, with PC failure occurring 50% higher; (11.9 ± 0.3)% for ER, compared to PC (17.9 ± 0.1)% (Table 2). Maximum longitudinal stress (σ_x^*) for PC was observed to be 256 ± 9 MPa, 24% greater than for ER (206 ± 2 MPa).

Fig. 4 displays the results from typical samples used to analyse the pseudo-ductile strain (ε_d) and the yield point (σ_{Yield}, $_{Yield}$). Included in the figure are the measurements taken. The results show a substantial change in slope after the matrix yields. The increasing stress beyond this point is believed to be mainly a result of fibre rotation [19]. No significant differences are observed post-matrix yielding for PC in comparison to ER, suggesting no strain hardening from the matrix. However, PC sustains greater ε_x, indicating that the high-strain matrix can be exploited in a $[\pm 45_n]_s$ configuration.

The averages of the measurements taken for all samples are shown in Table 2. Fig. 4 and the corresponding table highlight how the low yield point of ER and PC – together with the 45° angle plies - leads to a large pseudo-ductile strain. The yield stress was 36% greater for ER compared to PC (σ_{Yield} = 48.9 ± 0.3 MPa; $_{Yield}$ = 0.55 ± 0.01% for PC; σ_{Yield} = 66.3 ± 0.4 MPa; $_{Yield}$ = 0.55 ± 0.01% for ER), in addition, the elastic modulus was 35% greater for ER (E_x = 14.7 ± 0.3 GPa for ER; E_x = 10.9 ± 0.1 GPa for PC), similarly ER demonstrated a 28% higher shear chord modulus compared to PC (G_{12}^{chord} = 3.44 ± 0.03 GPa for ER; 2.68 ± 0.04 GPa for PC).

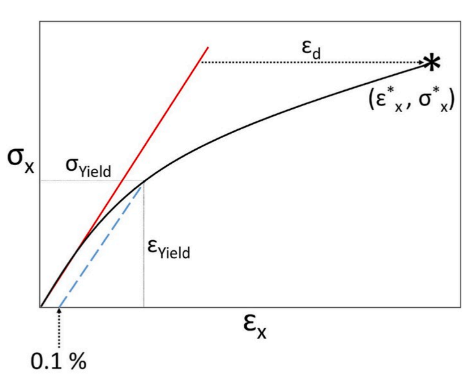

Fig. 2. Graphical explanation of the method used to determine the yield stress and pseudo-ductile strain.

However, PC demonstrated a 49% greater pseudo-ductile strain (ε_d = 15.6 ± 0.1% for PC; 11.9 ± 0.3% for ER) and PC demonstrated a 26% increase in apparent in-plane strength compared to ER (τ_{12}^m = 130.3 ± 4 MPa for PC; 103.2 ± 1 MPa for ER). In comparison, Herakovich et al. [20], reported with an IM7/K3B (polyimide matrix) an E_x of 15 GPa, and a σ_x^* of 375 MPa (compared to σ_x^* = 206 ± 2 MPa and 256 ± 9 MPa; E_x = 14.7 ± 0.3 GPa and 10.9 ± 0.1 GPa for ER and PC, respectively, reported in this work) and $_x$ of ~20% with [±45]. The greater elastic properties of the ER matrix led to superior σ_{Yield}, E_x and G_{12}^{chord}, however, the high strain characteristics of PC have benefited the composite properties of ε_d, σ_x^*, ε_x^* and τ_{12}^m.

PC samples were characterised by matrix cracking on the surface, consistent with Herakovich's findings [21]; this was not observed for ER; a comparison can be made between Fig. S4 in the supplementary information section of the PC sample prior to mechanical testing to Fig. 5b or Fig. S3 in the supplementary section of the PC sample after mechanical testing (note the change in the surface roughness).

As can be observed in Fig. 5 and verified by analysing the remaining samples, the delamination area on the edges for ER is greater than for PC).

As defined by ASTM D3518, 2007 [18] and quoted in the supplementary information (Table S1), failure area modes for the PC were: 3 × LG and 2 × LW; for ER are 1 × LW, 2 × AG and 2 × LG (where L is lateral failure type, A is angled failure type; G is gauge failure area and W is < 1 width from grip/tab failure area).

ER demonstrated a greater E_x, σ_{Yield} and G_{12}^{chord}, but PC demonstrated greater ε_d, ε_x^*, $\sigma_x^*$and τ_{12}^m. Evidently, the high strain-to-failure properties of PC with comparable tensile strength influenced the overall mechanical properties of the composite. It is expected that the inherent properties of polycarbonate, in comparison to a thermoset epoxy resin (which displays superior thermal stability, for instance), would ultimately dictate material selection in particular applications, but this study would be highly applicable where reinforced-thermoplastics are current employed (*e.g.* the automotive industry). A direct comparison of polycarbonate with a typical high-performance thermoset would be relatable to relevant industries and assists with potential material substitutions in the future.

No strain-hardening from the matrix was observed through comparing ER with PC post-matrix yield (Fig. 3); the non-linearity observed for both ER and PC is a consequence of fibre rotation [2]. Given the nature of the composite, it is unclear how the woven fibre configuration influences the formation of strain-bands under shear or if chain alignment is hindered. Strain hardening of an amorphous polymer, such as polycarbonate, is less pronounced in shear given the confinement of shear bands as a result of slender interlaminar regions and heterogeneous deformation [4,5,22,23]. Furthermore, thicker polymer samples display more strain hardening than thin samples [4]. The woven configuration adds complications when making comparisons with previous work. For example, Pozegic et al. [24] evaluated [±45] 2, 2 twill carbon fibre under in-plane shear loading, but the sample geometries were smaller than those in this work, which would be expected to create further confinement of fibre rotation; further work should assess the impact of this configuration during fibre rotation. It should be added that angle ply composites display a strain-rate sensitivity under tension, as the matrix becomes more significant than in unidirectional composites [ ] [25].

Future work should remove the contribution of the fibre rotation. This would provide a more accurate representation of τ_{12} and γ as can be reviewed in the following references: [2,19,20]; in doing so, consideration must be given to the variables: E_{22} and G_{12}, which are dependent on matrix and fibre-matrix interfacial properties and will change depending on the fibre rotation. The stress-strain response of polycarbonate (PC) is strain rate dependent [26], however strain-hardening has been observed in the neat polymer with the strain rates used in this

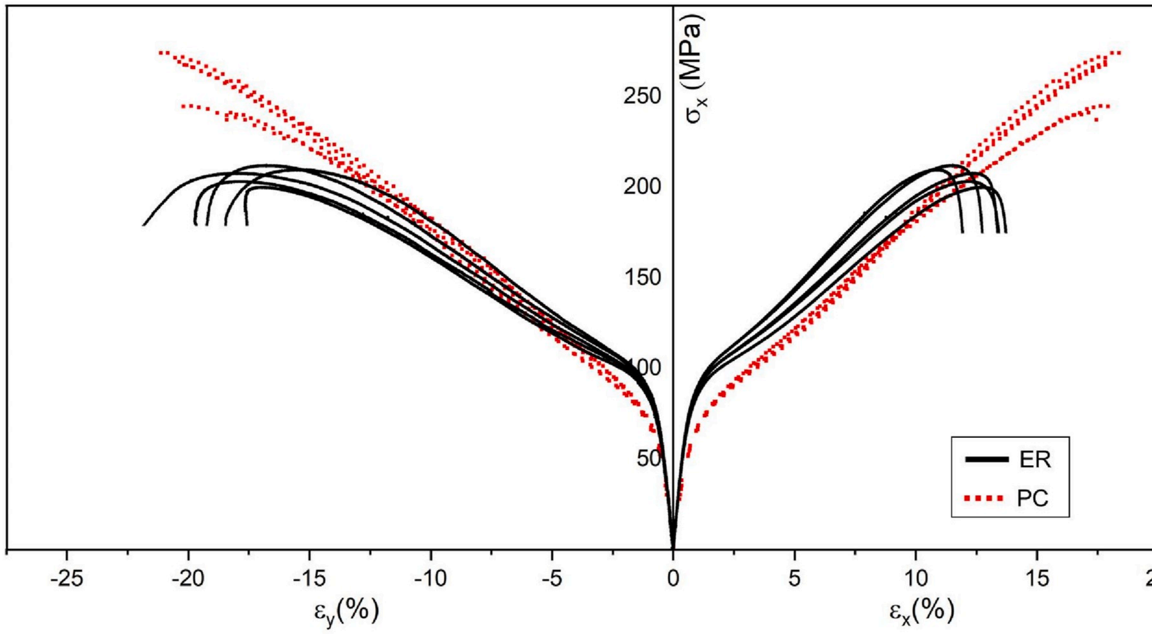

Fig. 3. *Applied longitudinal stress (σ_x) against longitudinal (ε_x) and transverse strains (ε_y) for ER (black, solid line) and PC (red, dashed line). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.).*

Table 2
Key parameters of the ER and PC samples.

Sample	E_x(GPa)	σ_{Yield} (MPa)	ε_{Yield}(%)	ε_d(%)	σ_x^*(MPa)	ε_x^*(%)	τ_{12}^m(MPa)	G_{12}^{chord}(GPa)
ER	14.7 ±0.3	66.3 ±0.4	0.55 ±0.01	10.5 ±0.3	206 ±2	11.9 ±0.3	103.2 ±1	3.44 ±0.03
PC	10.9 ±0.1	48.9 ±0.3	0.55 ±0.01	15.6 ±0.1	256 ±9	17.9 ±0.1	130.3 ±4	2.68 ±0.04

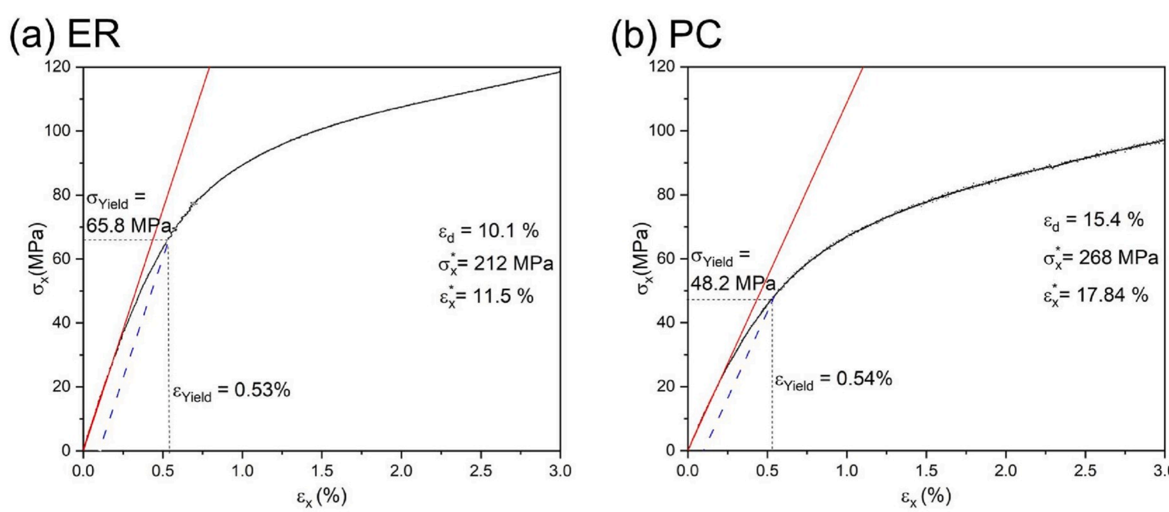

Fig. 4. Example of the pseudo-ductility at the yield point for (a) ER and (b) PC.

work. Nonetheless, testing fibre reinforced PC under different strain rates would be of interest for future work to determine rate dependence.

4. Conclusion

In developing pseudo-ductile composites through fibre rotation, the nature of the matrix was considered. Thus, polycarbonate matrix, which as a polymer, demonstrates strain-hardening and has a high strain-to-failure was compared to a commercial, epoxy resin matrix. The in-plane shear characteristics with shear strain were similar, however, the fibre reinforced polycarbonate composite demonstrated 49% improvement in pseudo-ductility and a 26% increase in apparent in-plane strength, at the cost, however, of modulus (−26% elastic and −22% shear chord modulus) and yield strength (−26%). Neither materials demonstrated premature matrix failure – before the development of non-linearity – as observed in UD $[\pm 45_n]_s$ composites with ply of

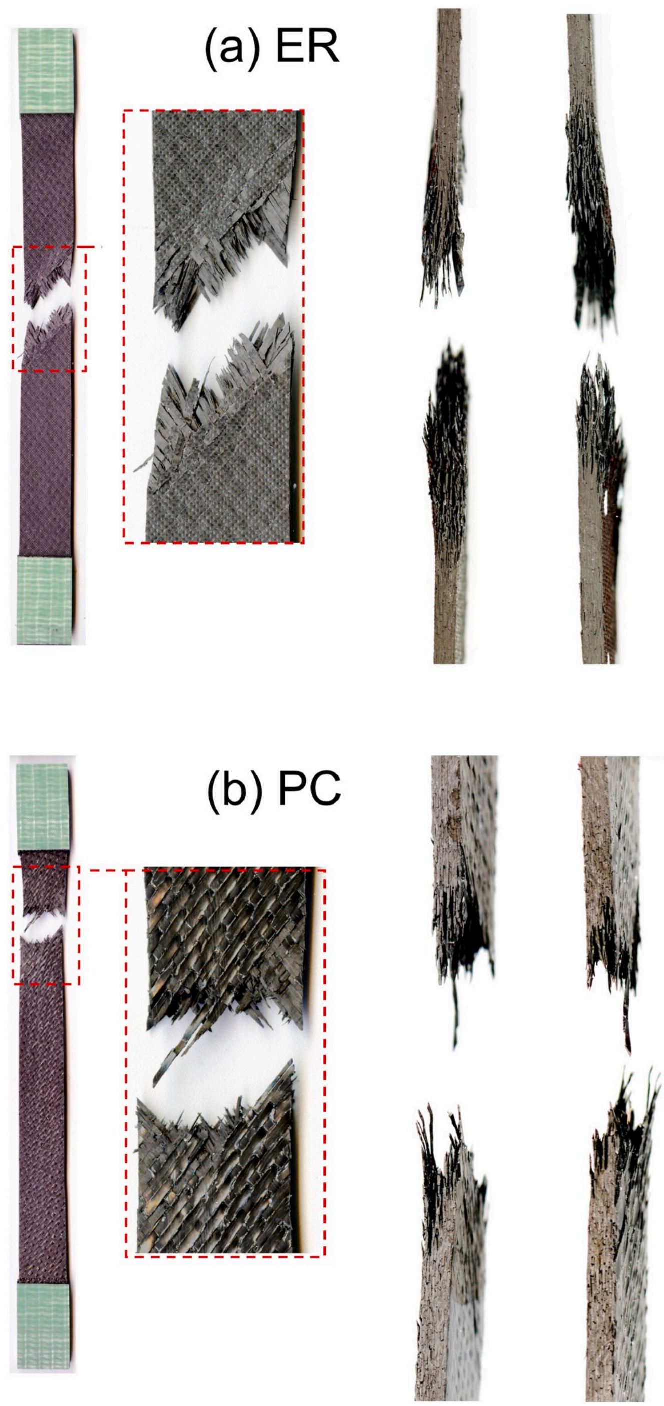

Fig. 5. Images of typical samples post-failure for (a) ER and (b) PC, with a higher magnification image of the failure area in the dashed red box and the sides of the specimens. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

thicknesses of >0.1 mm. Strain hardening was not observed post-matrix yield in PC, which could be attributed to strict confinement of the matrix during the entire elongation. Nevertheless, the thermoplastic polycarbonate demonstrates superior pseudo-ductile performance in terms of pseudo-ductile strain (49%), longitudinal strength (24%) and apparent in-plane shear strength (26%) to the thermoset epoxy resin matrix. Consequently, in applications where the thermal stability of thermoset matrices is not required, then the polycarbonate matrix may offer a worthy replacement.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Thomas R. Pozegic: Conceptualization, Methodology, Investigation, Data curation, Writing - original draft. **Mohamad Fotouhi:** Data curation, Writing - review & editing. **Xun Wu:** Data curation, Writing - review & editing. **Jamie W. Hartley:** Data curation, Writing - review & editing. **Michael R. Wisnom:** Writing - review & editing. **Ian Hamerton:** Project administration, Supervision, Data curation, Writing - review & editing.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.compscitech.2020.108261.

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